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(54) VANADIUM COMPLEX POSITIVE ELECTRODE FOR SOLID LITHIUM POLYMER BATTERY, AND LITHIUM POLYMER BATTERY USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a complex positive electrode for lithium polymer battery, capable of providing a small and light lithium polymer battery of high charging and discharging capacity and a lithium polymer battery, using the positive electrode.

SOLUTION: Macromolecule solid electrolyte having lithium salt as an electrolyte salt, solved in copolymer of average molecular weight higher one million (allyl glycidyl ether can be contained), composed of ethylene oxide, glycidyl ether including a side chain of ethylene oxide of degree of polymerization 1 to 12, a complex positive electrode composed of vanadium oxide and conductive particles or a complex positive electrode adding polyethylene glycol of average molecular weight 500 to 2,000 to the complex positive electrode mentioned above as a positive electrode active material are used. This lithium polymer battery is comprised of a negative electrode composed of lithium metal or lithium metal alloy, and a film with the macromolecule solid electrolyte made to bridge. The macromolecule solid electrolyte has lithium salt solved in copolymer of average molecular weight higher than one million composed of ethylene oxide, glycidyl ether including a side chain of ethylene oxide of degree of polymerization 1 to 12, and allyl glycidyl ether.

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CLAIMS

[Claim(s)]

[Claim 1] The compound positive electrode with which the weight average molecular weight which becomes 30-95 mol % and the side chain of ethylene oxide from 5-70 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12 is characterized by applying the vanadium system oxide VXO_5 ($X=2-2.5$) and the complex with which it consists of a conductive particle on a charge collector as the giant-molecule solid electrolyte which dissolved lithium salt in 1 million or more copolymers as an electrolyte salt, and a positive-active-material particle.

[Claim 2] The giant-molecule solid electrolyte with which the weight average molecular weight which becomes 30-95 mol % and the side chain of ethylene oxide from 5-70 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12 dissolved lithium salt in 1 million or more copolymers as an electrolyte salt, the compound positive electrode characterized by applying the complex with which a mean molecular weight consists of a vanadium system oxide VXO_5 ($X=2-2.5$) and a conductive particle as 500 or more polyethylene glycols [2000 or less] or the other compound of those, and a positive-active-material particle on a charge collector.

[Claim 3] The compound positive electrode with which the weight average molecular weight which becomes 30-94 mol % and the side chain of ethylene oxide from 5-69 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12, and 1-5 mol % of allyl glycidyl ether is characterized by applying the vanadium system oxide VXO_5 ($X=2-2.5$) and the complex with which it consists of a conductive particle on a charge collector as the giant-molecule solid electrolyte which dissolved lithium salt in 1 million or more copolymers as an electrolyte salt, and a positive-active-material particle.

[Claim 4] 30-94 mol % of ethylene oxide, The solid polymer electrolyte with which the weight average molecular weight which becomes a side chain from 5-69 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12, and 1-5 mol % of allyl glycidyl ether dissolved lithium salt in 1 million or more copolymers as an electrolyte salt, The compound positive electrode characterized by applying the complex with which weight average molecular weight consists of a vanadium system oxide VXO₅ (X=2-2.5) and a conductive particle as 500 or more polyethylene glycols [2000 or less] or the ether compound of those, and a positive-active-material particle on a charge collector.

[Claim 5] The compound positive electrode according to claim 3 or 4 characterized by making the solid polymer electrolyte in a compound positive electrode construct a bridge.

[Claim 6] the lithium salt used for a giant-molecule solid electrolyte -- LiN(CF₃SO₂)₂ -- it is -- this lithium salt -- the copolymer 100 weight section -- receiving -- **** for 5 - 50 weight sections -- a compound positive electrode given in either of claim 1 to claims 5 characterized by things.

[Claim 7] a conductive particle -- KETCHIEN black or acetylene black -- it is -- these conductive particles -- the positive-active-material particle 100 weight section -- receiving -- **** for 5 - 20 weight sections -- a compound positive electrode given in either of claim 1 to claims 5 characterized by things.

[Claim 8] the positive-active-material particle 100 weight section -- receiving -- a solid polymer electrolyte 5 - **** for 35 weight sections -- a compound positive electrode given in either of claim 1 to claims 5 characterized by things.

[Claim 9] The lithium-polymer battery with which the weight average molecular weight which becomes the negative electrode which becomes either of claim 1 to claims 5 from the compound positive electrode, lithium metal, or lithium metal alloy of a publication, and 30-94 mol % and the side chain of ethylene oxide from 5-69 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12, and 1-5 mol % of allyl glycidyl ether consists of film over which the giant-molecule solid electrolyte which dissolved lithium salt in 1 million or more copolymers was made to construct a bridge.

[Claim 10] The charge approach of the lithium-polymer battery characterized by ending in 3.6-4.2V in the charge approach of a lithium-polymer battery according to claim 9.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the compound positive electrode for lithium-polymer batteries, the lithium-polymer battery which used the positive electrode, and its operation.

[0002]

[Description of the Prior Art] About the lithium secondary battery, research is made actively recently and many proposals are made about the cell component or the assembly. For example, LiCoO₂, LiNiO₂, LiMn₂O₄, V₂O₅, V₆O₁₃, and TiS₂ grade are used as positive active material, and the rechargeable battery using a lithium and lithium-

aluminium alloy, carbon (hard carbon, a natural graphite, a mesophase carbon micro bead, mesophase carbon fiber), etc. as a negative-electrode active material is proposed. In these lithium secondary batteries, the electrolytic solution made to dissolve the lithium salt of LiClO_4 , LiBF_4 , LiAsF_6 , LiPF_6 , LiCF_3SO_3 , and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ grade in one or more sorts of aprotic organic solvents, such as propylene carbonate [which a lithium ion can move], ethylene carbonate, diethyl carbonate, 1, and 2-dimethoxyethane, as the electrolytic solution is used. However, since these electrolytic solutions are inflammability, the lithium secondary battery which used these electrolytic solutions has the danger of ignition or explosion. Moreover, when a lithium metal and a lithium alloy are used as a negative electrode, there is a danger that the lithium dent light generated on a negative electrode will reach and connect with a positive electrode too hastily. It originates in these troubles using the organic solvent for the electrolytic solution. [0003] Although development of the lithium-polymer battery which gelled the electrolytic solution by the polymer and was fixed is furthered in order to solve these troubles, the use under the problem of a liquid spill of the electrolytic solution or hot environments 60 degrees C or more is not borne yet. Then, research of the solid-state mold lithium cell using a perfect solid electrolyte polymer is advanced. It is J.Electrochem.Soc., 147, and 2050 (2000) that the cell property in which the lithium-polymer battery which combined the positive electrode which used the molecular-weight about 2000 polyethylene glycol as a binder-cum-an ionic conductor of a positive electrode, the giant-molecule solid electrolyte, and the lithium metal was excellent in 60 degrees C with summer is shown. It is reported. However, especially the lithium-polymer battery that used this molecular-weight about 2000 polyethylene glycol for the positive electrode has more the fault in which the cycle property of a cell deteriorates greatly at the elevated temperature (80 degrees C or more). Moreover, since this polyethylene glycol is a liquid, it is necessary to add so much in a positive electrode, and that amount is required [binding capacity with positive active material and an electric conduction agent is weak under an elevated temperature 60 degrees C or more, and] for it more than 50 weight sections to the positive-active-material particle 100 weight section. For this reason, the rate of the active material in a compound positive electrode will decrease, and the energy density which a compound positive electrode has will become small. The positive electrode for lithium-polymer batteries which can still be equal to practical use for a such fault is not developed. Moreover, in Japanese Patent Application No. 10-85890, the patent about the positive electrode for lithium-polymer batteries using V2O5 as positive active material is described. The cell using the positive electrode which used V2O5 as the active material in this patent has an upper limit electrical potential difference as low as 3.0V at the time of charge, and the discharge capacity of the cell at this time is small as a cell which used V2O5.

[0004]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the compound positive electrode for lithium-polymer batteries which can offer a lithium-polymer battery with a large charge-and-discharge capacity by the small light weight, and the lithium-polymer battery using the positive electrode.

[0005]

[Means for Solving the Problem] Although 3.5 V was an upper limit conventionally when it was a banazin san ghost, although it is more desirable to be able to charge by the high

voltage in order to obtain a lithium-polymer battery with a large charge-and-discharge capacity by the small light weight, the compound positive electrode which can be charged to 4.2 V was found out.

[0006] That is, this inventions are the vanadium system oxide VXO_5 ($X=2-2.5$) and a compound positive electrode with which it is characterized by applying the complex with which it consists of a conductive particle on a charge collector as the giant-molecule solid electrolyte with which the weight average molecular weight which becomes 30-95 mol % and the side chain of ethylene oxide from 5-70 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12 dissolved lithium salt in 1 million or more copolymers as an electrolyte salt, and a positive-active-material particle. Furthermore, this invention offers the lithium-polymer battery with which the weight average molecular weight which becomes the negative electrode which consists of the above-mentioned compound positive electrode and a lithium metal, or a lithium metal alloy, and 30-94 mol % and the side chain of ethylene oxide from 5-69 mol % of glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12, and 1-5 mol % of allyl glycidyl ether consists of film over which the giant-molecule solid electrolyte which dissolved lithium salt in 1 million or more copolymers was made to construct a bridge, and offers the charge approach.

[0007] The giant-molecule solid electrolyte with desirable using it in the compound positive electrode in this invention is a solid polymer electrolyte with which the weight average molecular weight which becomes ethylene oxide and a side chain from the glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12 added lithium salt as an electrolyte salt to 1 million or more copolymers. Compared with the molecular-weight about 2000 polyethylene glycol, since molecular weight is high, this solid electrolyte is excellent in the mechanical strength. Since the melting point is still higher, it excels in the binding property with a positive-active-material particle and a conductive particle under the elevated temperature 60 degrees C or more. By using this solid polymer electrolyte excellent in the binding property for a compound positive electrode, the amount of the solid polymer electrolyte in a compound positive electrode can be lessened. Thereby, the energy density of a compound positive electrode can be raised. Moreover, to the bottom of an elevated temperature 80 degrees C or more, since it is a solid-state, compared with the compound positive electrode produced using the mean-molecular-weight about 2000 polyethylene glycol, degradation of a positive electrode cannot take place easily. From this, the lithium-polymer battery of the positive electrode using this giant molecule can demonstrate not only 60 degrees C but the still higher cell engine performance also under an elevated temperature (80 degrees C or more). In this invention, a compound positive electrode is produced by applying the complex of a solid polymer electrolyte, a positive-active-material particle, and a conductive particle on a charge collector. The thing of configurations, such as a foil which consists of aluminum, platinum, and carbon as a charge collector, a mesh, and foaming, can be used.

[0008] By making a bridge construct, the solid polymer electrolyte which made the copolymer which carried out copolymerization of the allyl glycidyl ether to the glycidyl ether which has the ethylene oxide unit of degrees of polymerization 1-12 further dissolve lithium salt in ethylene oxide and a side chain as an electrolyte salt can raise the reinforcement of a solid polymer electrolyte further. For this reason, after applying the

complex of this solid polymer electrolyte, a positive-active-material particle, and a conductive particle on a charge collector, if bridge formation is performed, it is possible to raise further a binding property with a solid polymer electrolyte, a positive-active-material particle, and a conductive particle. As the bridge formation approach at this time, activity energy lines, such as a radical initiator chosen from organic peroxide, an azo compound, etc., ultraviolet rays, and an electron ray, can be used. A mean molecular weight can add 500 or more polyethylene glycols [2000 or less] or the ether compound of those in a compound positive electrode. As an ether compound of a polyethylene glycol, monochrome or wood ether and monochrome, or diethylether is good. By constructing a bridge in the solid polymer electrolyte in a compound positive electrode, a binding property with a positive-active-material particle and a conductive particle becomes strong. For this reason, even if it adds the polyethylene glycol inferior to a binding property, or its ether compound in a compound positive electrode, degradation of the positive electrode by repeating a charge-and-discharge cycle can be decreased. Moreover, since 500 or more polyethylene glycols [2000 or less] or the ether compound of those excels [mean molecular weight] in the ion conductivity under an elevated temperature (60 degrees C or more), if it adds in a positive electrode, it will become possible to improve the charge-and-discharge property of the cell especially at the time of a high current.

[0009] As a 3 Class V positive-electrode ingredient of a lithium polymer rechargeable battery, the vanadium system oxide has been used widely. Cell voltage also increases a vanadium system oxide in monotone as charge progresses. Although a lithium-polymer battery uses a solid polymer electrolyte as an electrolyte, it is inferior to ion conductivity compared with the organic electrolytic solution, and its polarization of the electrical potential difference at the time of charge and discharge is large. For this reason, the capacity which can be charged will decrease by polarization at the time of charge. When a solid polymer electrolyte decomposed by the cell using polyethylene oxide as a conventional giant-molecule solid electrolyte, the limitation of a charge electrical potential difference was 3.5V. Moreover, also in the patent (Japanese Patent Application No. 10-85890) mentioned above, the upper limit of a charge electrical potential difference was 3.0V. The ethylene oxide in this invention (30-94-mol %), The weight average molecular weight which becomes a side chain from the glycidyl ether (5-69-mol %) which has the ethylene oxide unit of degrees of polymerization 1-12, and allyl glycidyl ether (1-5-mol %) to 1 million or more copolymers as an electrolyte salt The lithium-polymer battery which used the film over which the giant-molecule solid electrolyte which dissolved lithium salt was made to construct a bridge as a giant-molecule solid-electrolyte membrane can be charged more to the high voltage (4.2V), and can be used as a cell [high capacity / lithium-polymer battery / conventional / by this]. Moreover, since an electrical potential difference goes abruptly up at the time of a full charge, charge control can be easily performed by setting up a charge termination electrical potential difference.

[0010] As an electrolyte salt, the lithium salt of LiClO_4 , LiBF_4 , LiAsF_6 , LiPF_6 , LiCF_3SO_3 , and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ grade can be used. As lithium salt used for the giant-molecule solid electrolyte in a positive electrode, $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ can use it simpler among these. This is based on the following reasons. It hydrolyzes easily with moisture and HF generates lithium salt like LiPF_6 currently widely used for the lithium secondary

battery. This generated HF will disassemble a solid polymer electrolyte. Or it reacts with a lithium metal and lithium fluoride is generated. On the other hand, hydrolysis according [$\text{LiN}(\text{CF}_3\text{SO}_2)_2$] to moisture is because there is nothing and HF is hardly generated, either. Moreover, if there are few amounts of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ added to the copolymer 100 weight section than 5 weight sections, the concentration of the lithium ion in a giant-molecule solid electrolyte will be too thin, and sufficient lithium ion conductivity will not be acquired. Moreover, to the copolymer 100 weight section, lithium salt cannot finish melting into a copolymer and more than 50 weight sections cannot be added. As lithium salt in [this to] a giant-molecule solid electrolyte, $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ is good, and the amount has good 5 - 50 weight section to the copolymer 100 weight section.

[0011] There is no electronic conduction nature in the positive-active-material particle contained in a compound positive electrode, and a solid polymer electrolyte. So, in order for an oxidation reduction reaction to start in the positive-active-material particle in a compound positive electrode, it becomes indispensable to add a conductive particle into a compound positive electrode. KETCHIEN Black and acetylene black which give high electronic conduction nature to a compound positive electrode with an addition small as a conductive particle are suitable. The conductive particle in a compound positive electrode has good 5 - 20 weight section to the positive-active-material 100 weight section. To the positive-active-material particle 100 weight section, if this has few additions of a conductive particle than 5 weight sections, it cannot give sufficient electronic conductivity for a compound positive electrode, and the oxidation reduction reaction in the positive-active-material particle in a compound positive electrode does not fully occur. Thereby, the utilization factor of a positive-active-material particle falls, and charge-and-discharge capacity decreases more greatly than a theoretical value. On the other hand, if it adds more than 20 weight sections to the positive-active-material particle 100 weight section, although the electronic conductivity of a compound positive electrode is securable, the rate of the positive active material in a compound positive electrode falls. Thereby, the capacity of a compound positive electrode will fall.

[0012] the inside of a compound positive electrode -- the positive-active-material particle 100 weight section -- receiving -- a solid polymer electrolyte -- 5 - 35 weight **** rare ***** -- things are good. Binding capacity [as opposed to / that solid polymer electrolytes are below 5 weight sections / a positive-active-material particle] is weak, and this cannot produce a compound positive electrode. Moreover, the positive-active-material particle in a compound positive electrode of what has enough binding capacity will decrease that they are more than 35 weight sections, and the capacity of a compound positive electrode will fall. The weight average molecular weight of the copolymer used by this example computed molecular weight by standard polystyrene conversion with the gel par MYUESHON chromatography measuring method. Gel par MYUESHON chromatography measurement was performed at 60 degrees C using Shodex KD-806 of measuring device RID-6A of Shimadzu Corp., and the column by Showa Denko K.K., KD-806M, KD-803, and Solvent DMF.

[0013]

[Example] Hereafter, an example is shown and this invention is explained concretely. 2Oexample 1V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, it is the copolymer (weight average molecular weight: 1,500,000) of ethylene oxide (88-mol %) and 2-(2-methoxyethoxy)

ethyl glycidyl ether (12-mol %). 0.15 g and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.033g were dissolved in the acetonitrile. The above-mentioned solution was added to V2O5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the thickness of the whole positive electrode produced the compound positive electrode which is 40 micrometers. The obtained compound positive electrode, As solid polymer electrolyte film The copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) is made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$. Bridge formation processing Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the performed solid polymer electrolyte to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the 60-degree C thermostat, constant-current charge and 0.2C were performed by 0.5C and upper limit electrical-potential-difference 4.1V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. The discharge capacity when repeating charge and discharge is shown in drawing 1. This drawing shows that a high initial discharge capacity of about 300 mAh(s) is shown per 1g of positive active material (mAh/g expresses the charge-and-discharge capacity per 1g of positive active material henceforth). 80 cycles after was found by that the cell which was maintaining as high a discharge capacity as 200 mAh/g, and made it as an experiment shows a high cycle property in 60 degrees C.

[0014] 2Oexample 2V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, 0.20g (weight average molecular weight: 1,500,000) of copolymers of ethylene oxide (88-mol %) and 2-(2-methoxyethoxy) ethyl glycidyl ether (12-mol %) and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.067g were dissolved in the acetonitrile. The above-mentioned solution was added to V2O5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The obtained compound positive electrode, As solid polymer electrolyte film The copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) is made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$. Bridge formation processing Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the performed solid polymer electrolyte to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the thermostat (60 degrees C and 80 degrees C), constant-current charge and 0.2C were performed by 0.2C and upper limit electrical-potential-difference 4.1V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. The cell test temperature showed the same high cycle property as the time of 60 degrees C also in 80 degrees C.

[0015] 2Oexample of comparison 1V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.13g and often mixed with the mortar. On the other hand, it is the polyethylene-glycol monomethyl ether of a mean molecular weight 2000 [about]. 0.53g

and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.177g were dissolved in the acetonitrile. The above-mentioned solution was added to V_2O_5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The obtained compound positive electrode, As solid polymer electrolyte film The copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) is made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$. Bridge formation processing Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the performed solid polymer electrolyte to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the thermostat (60 degrees C and 80 degrees C), constant-current charge and 0.2C were performed by 0.2C and upper limit electrical-potential-difference 4.1V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. In 80 degrees C, degradation of a cycle property became [the cell test temperature] large compared with the time of 60 degrees C.

[0016]

[Table 1]

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[0017] 2Oexample 3V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, 0.15g (weight average molecular weight: 1,500,000) of copolymers of ethylene oxide (88-mol %) and 2-(2-methoxyethoxy) ethyl glycidyl ether (12-mol %) and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.05g were dissolved in the acetonitrile. The above-mentioned solution was added to V_2O_5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The obtained compound positive electrode, As solid polymer electrolyte film The copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) is made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$. Bridge formation processing Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the performed solid polymer electrolyte to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. 0.2C and an upper limit electrical potential difference were changed with 3.2, 3.5, 3.8, 4.1, and 4.4V in the 60-degree C thermostat, and constant-current discharge was performed by constant-current charge, 0.2C, and minimum electrical-potential-difference 2.0V. The upper limit electrical potential difference was able to charge satisfactory more than 3.6V. The discharge capacity in each upper limit electrical potential difference is shown in drawing 2. The discharge capacity of a cell was able to be made to increase by

from now on charging to a high electrical potential difference. The relation of the charge-and-discharge capacity and cell voltage of 10 cycle eye is shown for a charge-and-discharge cycle in drawing 3. It turns out that cell voltage is going abruptly up near capacity 270 mAh/g which hits a full charge from this drawing. By setting a charge termination electrical potential difference as 4.0-4.2V showed that charge was easily controllable.

[0018] 2Oexample 4V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, 0.10g (weight average molecular weight: 1,500,000) of copolymers of ethylene oxide (88-mol %) and 2-(2-methoxyethoxy) ethyl glycidyl ether (12-mol %) and $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.033g were dissolved in the acetonitrile. The above-mentioned solution was added to V_2O_5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The obtained compound positive electrode, As solid polymer electrolyte film The copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) is made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$. Bridge formation processing Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the performed solid polymer electrolyte to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the 60-degree C thermostat, constant-current charge was performed by upper limit electrical-potential-difference 4.1 V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. 0. The capacity of a cell turned into 76% of initial capacity in charge and discharge with the current of 2C at the time of 200 cycle *****. Moreover, when charge and discharge were performed by 0.5C, it became about 68% of the capacity when performing charge and discharge by 0.2C.

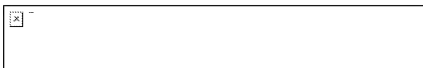
[0019] 2Oexample 5V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, 0.10g (weight average molecular weight: 1,500,000) of copolymers of ethylene oxide (82-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (18-mol %), and allyl glycidyl ether (1.7-mol %), $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ 0.033g, and 0.005g of benzoyl peroxides were dissolved in the acetonitrile. The above-mentioned solution was added to V_2O_5 and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The bridge was constructed in the polymer in a compound positive electrode by performing 100 degrees C and 3-hour heat-treatment for this compound positive electrode among argon gas. Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the solid polymer electrolyte which the copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) was made to carry out 30 weight sections dissolution of $\text{LiN}(\text{CF}_3\text{SO}_2)_2$, and performed bridge formation processing in it

as a giant-molecule solid-electrolyte membrane to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the 60-degree C thermostat, constant-current charge was performed by upper limit electrical-potential-difference 4.1V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. 0. The capacity of a cell turned into 81% of initial capacity in charge and discharge with the current of 2C at the time of 200 cycle *****. Moreover, when charge and discharge were performed by 0.5C, it became about 63% of the capacity when performing charge and discharge by 0.2C. Since the binding property with a positive-active-material particle and a conductive particle went up this by carrying out bridge formation processing of the polymer in a compound positive electrode, since the ion conductivity of the solid polymer electrolyte in a compound positive electrode fell by elongation and bridge formation processing, the cycle life of a cell is considered that the fall of the capacity in 0.5C was large.

[0020] 2Oexample 6V5 powder 1.0g, KETCHIEN Black It extracted by the ratio of 0.15g and often mixed with the mortar. On the other hand, they are ethylene oxide (82-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (18-mol %), and 0.10g (weight average molecular weight: 1,500,000) of copolymers of allyl glycidyl ether (1.7-mol %) and the polyethylene-glycol wood ether of molecular weight 2000 [about]. 0.05g, LiN(CF₃SO₂)₂ 0.033g, benzoyl peroxide 0.005g was dissolved in the acetonitrile. The above-mentioned solution was added to V2O₅ and KETCHIEN Black mixing powder, it often mixed with the mortar, and the positive-electrode slurry was obtained. After applying this slurry to aluminum foil with a thickness of 20 micrometers, the solvent was removed in the 80-degree C oven. By carrying out the roll press of this, the compound positive electrode with a thickness [of the whole positive electrode] of 40 micrometers was produced. The bridge was constructed in the polymer in a compound positive electrode by performing 100 degrees C and 3-hour heat-treatment for this compound positive electrode among argon gas. Li metallic foil with a thickness of 100 micrometers was made to rival as what produced the solid polymer electrolyte which the copolymer 100 weight section of ethylene oxide (80.6-mol %), 2-(2-methoxyethoxy) ethyl glycidyl ether (17.7-mol %), and allyl glycidyl ether (1.7-mol %) was made to carry out 30 weight sections dissolution of LiN(CF₃SO₂)₂, and performed bridge formation processing in it as a giant-molecule solid-electrolyte membrane to 50 micrometers of thickness, and a negative electrode, and the coin mold cel was produced. In the 60-degree C thermostat, constant-current charge was performed by upper limit electrical-potential-difference 4.1V, and constant-current discharge was performed by minimum electrical-potential-difference 2.0V. 0. The capacity of a cell turned into 79% of initial capacity in charge and discharge with the current of 2C at the time of 200 cycle *****. Moreover, when charge and discharge were performed by 0.5C, it became about 72% of the capacity when performing charge and discharge by 0.2C. This is considered that the fall of the capacity in 0.5C became small by the polyethylene-glycol wood ether which the cycle life of a cell added in elongation and a compound positive electrode since the binding property with a positive-active-material particle and a conductive particle went up since ion conductivity improved by carrying out bridge formation processing of the polymer in a compound positive electrode.

[0021]

[Table 2]



[0022]

[Effect of the Invention] According to this invention, the compound positive electrode which offers a lithium-polymer battery with a large charge-and-discharge capacity by the small light weight, and the lithium-polymer battery using the positive electrode are obtained. The cell of this invention can be charged to a high electrical potential difference, and can attain high capacity by this. Since the cell of this invention operates to stability under an elevated temperature, it can be used as the electric vehicle which cell temperature tends to go up, a hybrid car, a cell for load leveling, etc.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] The relation of the discharge capacity of all solid-state lithium-polymer batteries and the cycle property of having used the V2O5 compound positive electrode is shown.

[Drawing 2] The relation between the upper limit electrical potential difference at the time of charge of the lithium-polymer battery using a V2O5 compound positive electrode and discharge capacity is shown.

[Drawing 3] The relation of the charge-and-discharge capacity and cell voltage of the lithium-polymer battery using a V2O5 compound positive electrode is shown.
